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Measurement of ground state properties of neutron-rich nuclei on the r-process path between the N=50 and N=82 shells

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Abstract

The evolution of the unknown ground-state β-decay properties of the neutron-rich $^{84-89}$Ge, $^{90-93}$Se and $^{102-104}$Sr isotopes near the r-process path is of high interest for the study of the abundance peaks around the N=50 and N=82 neutron shells. At ISOLDE, beams of certain elements with sufficient isotopic purity are produced as molecular sidebands rather than atomic beams. This applies e.g. to germanium, separated as GeS^+, selenium separated as $SeCO^+$ and strontium separated as SrF^+. However, in case of neutron-rich isotopes produced in actinide targets, new “isobaric” background of atomic ions appears on the mass of the molecular sideband.

For this particular case, the ECR charge breeder, positioned in the experimental hall after ISOLDE first mass separation, can be advantageously used as a purification device, by breaking the molecules and removing the molecular contaminants.

This proposal indicates our interest in the study of basic nuclear structure properties of neutron-rich nuclei on the r-process path between the neutron magic numbers N=50 and N=82 at ISOLDE with the ECR Phoenix Booster. This first proposal should serve as a basis for the investigation of the potential applications of the ECR for nuclear astrophysics at ISOLDE. For instance, beam purification using molecular sidebands selection with the ECR is particularly suited for the study of the $^{84-89}$Ge, $^{90-93}$Se, $^{102-104}$Sr isotopes, respectively produced as GeS^+, $SeCO^+$ and SrF^+ molecular beams.

1. Motivation

We propose to determine the half lives and Pn-values of neutron rich isotopes located close to the r-process path between shells N=50 and N=82.

1.1. Astrophysics case

Approximately half of the heaviest isotopes in nature, beyond iron, are produced via neutron captures on very short time scales in neutron-rich environments, i.e. the so-called r-process.
At the high temperatures and neutron densities present in the r-process site, equilibrium between neutron captures and photo-dissociations is achieved. For each isotopic chain the series of neutron captures reaches equilibrium with photo-dissociation in what is known as “waiting point” nucleus as the flow of neutron captures "waits" for this nucleus to beta-decay. The r-process reaches the neutron shell closures at N = 50, 82, and 126 at such low Z values that the neutron separation energy is too small to allow the formation of still more neutron-rich isotopes; the isotopes then have to beta decay. To overcome the shell gap at the magic neutron numbers and produce heavier nuclei, the material has to undergo a series of alternating β-decays and neutron captures before it reaches a nucleus close enough to stability to have neutron separation energies large enough to allow for the continuation of the sequence of neutron capture reactions. As the β-decay half-lives are relatively long at the magic neutron numbers, the r-process network waits long enough at these neutron numbers to build up abundance peaks related to the mass numbers A = 80, 130, and 195. These abundance peaks are thus mainly shaped by the β-decay properties of the nuclei on the r-process path located between the neutron closed shells. Presently, most of these properties are not experimentally measured.

1.2. Comparison with models

The beta-decay half-lives ($T_{1/2}$) and the β-delayed neutron emission probabilities (P_n) combined provide a first rough characterization of the distribution of beta-strength in a beta-decay. In nuclei with N > 50 around and above Ni, first forbidden transitions will play an important role [1] and affect both experimental quantities. The correct theoretical description of these nuclei is an ongoing project (see for example [2], for a recent review).

2. Description of the PHOENIX ECR Booster

At ISOLDE, neutron-rich isotopes in the intermediate mass range (A=80 to 150) can be produced with high yields by fission of an actinide target associated to a chemically non-selective [3] plasma ion source. Thus, the whole isobaric chain is ionized, resulting sometimes in a high background of many different fission products that are extracted and mass-separated. This kind of problem can be addressed using an electron cyclotron ion source (ECRIS) to charge breed and remove isobaric contaminants with one of the beam purification techniques described later in this section.

The Daresbury PHOENIX ECRIS Booster is installed after the General Purpose Separator (GPS), on the High Mass (GHM) parasitic beam line at ISOLDE (see Fig.1) as an on-line test bench for charge breeding experiments described in a previous proposal [4].

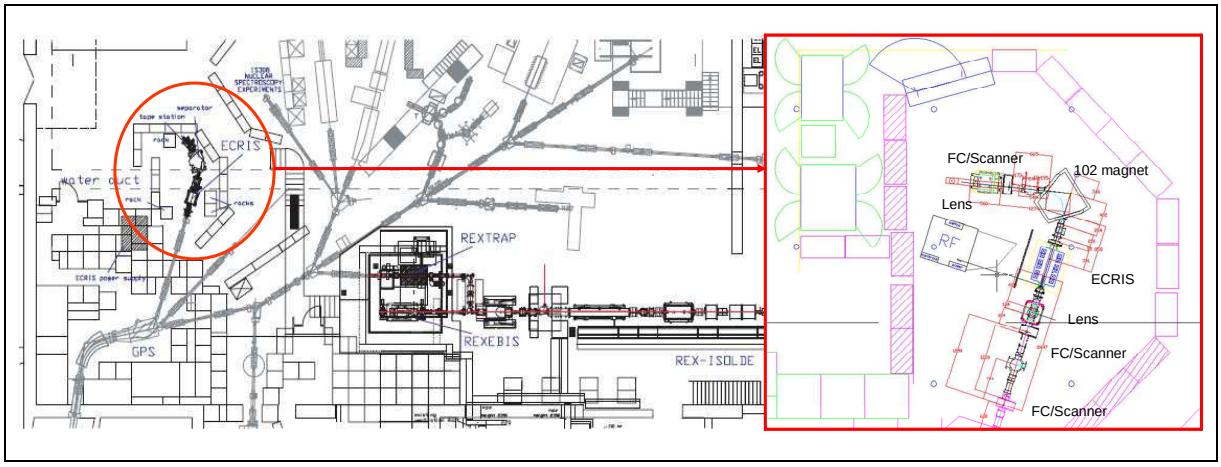

Fig.1 Current layout of the PHOENIX Booster ECRIS in ISOLDE hall

The singly charged ions from ISOLDE are injected into the PHOENIX Booster plasma chamber where they are slowed down by the ECRIS platform potential and stopped due to Coulomb interaction with the plasma (see Fig.2). The electrons of the plasma are confined in a so-called minimum-B-structure. A closed surface is created where the electron cyclotron resonance condition is fulfilled. The high mirror ratio of the magnetic field leads to long confinement times for the plasma electrons. They can pass thousands times the resonance region, stochastically gaining energy from a high frequency electromagnetic wave and ionizing plasma ions to high charge states via stepwise ionization.

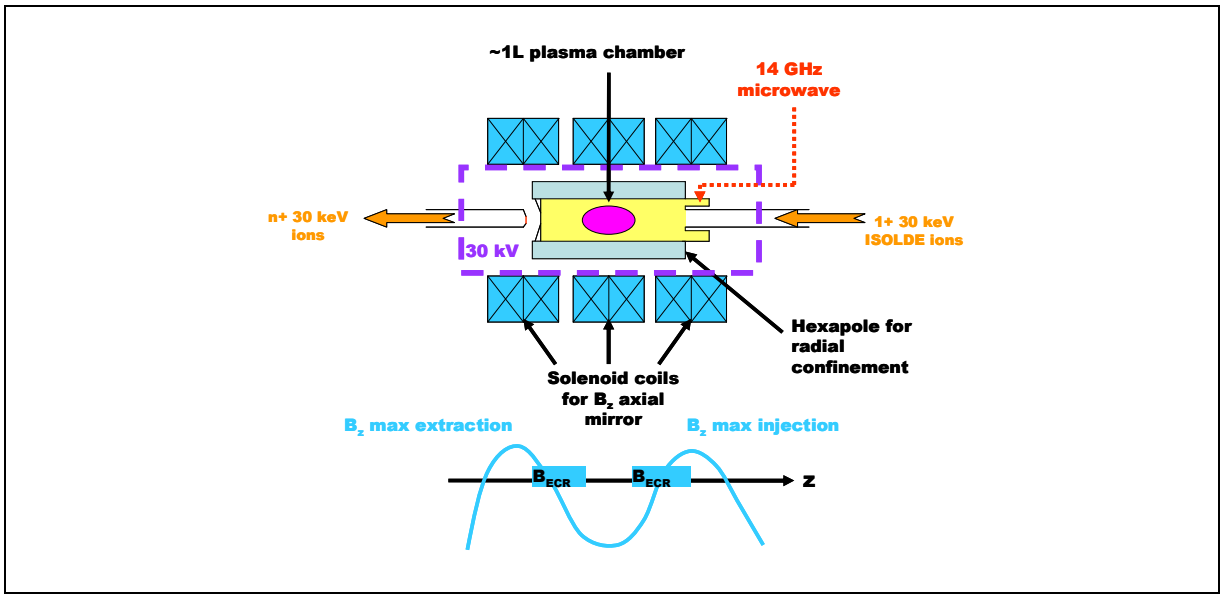

Fig.2 Ion motion inside the ECRIS

The latest on-line charge breeding results of the Phoenix booster are described in [5]. It is currently able to charge breed a wide range of nuclides for A/q values from 4 to 8, either in continuous or in pulsed mode. Efficiencies of the injected elements are similar to the ones measured for the REX beam preparation stage [6]. Charge bred ions are extracted and mass-separated by the 102º analyzing magnet (see Fig.3) and can be sent towards a tape station around which β and γ detectors are set up.

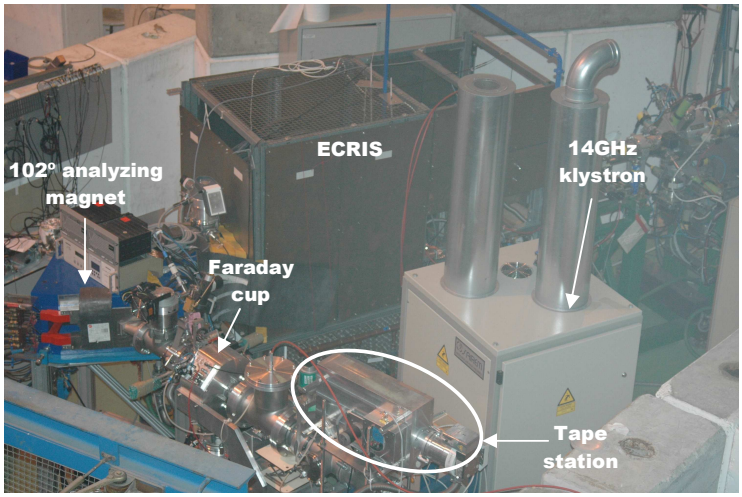

Fig.3 ECRIS experimental area

The PHOENIX Booster charge breeder offers two means for beam purification. The first one is the use of the charge state distribution generated inside the ECR plasma to extract ions with an optimized A/q-value so that the proportion of multiply charged contaminants is strongly suppressed. This method was already applied at ISOLDE to perform the study of ^{48}Ar decay [7].
The second method is the injection of the wanted species as molecules into the charge breeder where they are broken, extracted and mass selected, thus removing the molecular sidebands. Some elements are extracted from the ISOLDE target with a higher yield and less contamination as molecular beams.

As mentioned before, the question of isobaric contamination is crucial for the proper measurement of nuclear structure properties. As an example, the case of neutron-rich Ga was already studied at ISOLDE by K.-L. Kratz et al [8], where a ^{238}UC-graphite target was connected to a plasma ion source to produce the ^{81,82,83,84}Ga isotopes. According to reference [8], "*The determination of the Pn-values of the isotopes of major astrophysical interest was difficult due to the chemical nonselectivity of the plasma ion source. […] Therefore, the Pn-values listed (in Tab. 1) have rather large uncertainties"*. Now, a resonant laser ion source can be used at ISOLDE for selective ionization of many metallic elements. However, many non-metallic and some metallic elements can be better (i.e. more efficiently and/or more purely) separated in molecular form with a plasma ion source. The PHOENIX Booster charge breeding thus appears as an advantageous solution for cases where molecular sidebands can be injected.

3. Performances of the charge breeder

The PHOENIX Booster ECR charge breeder performances were studied in a previous proposal [4]. Charge breeding can be achieved within a continuous mode or a pulsed mode.

In continuous mode, the radiofrequency is applied permanently and the charge bred ions are extracted continuously from the source. The total time spent by the ions inside the plasma chamber is a global confinement time which includes the needed time for charge breeding [5]. Depending on the adjustments of the source, the global confinement time can go down to an average value of 100ms. Working in continuous mode, a charge breeding efficiency of typically 5% can be expected for one charge state.

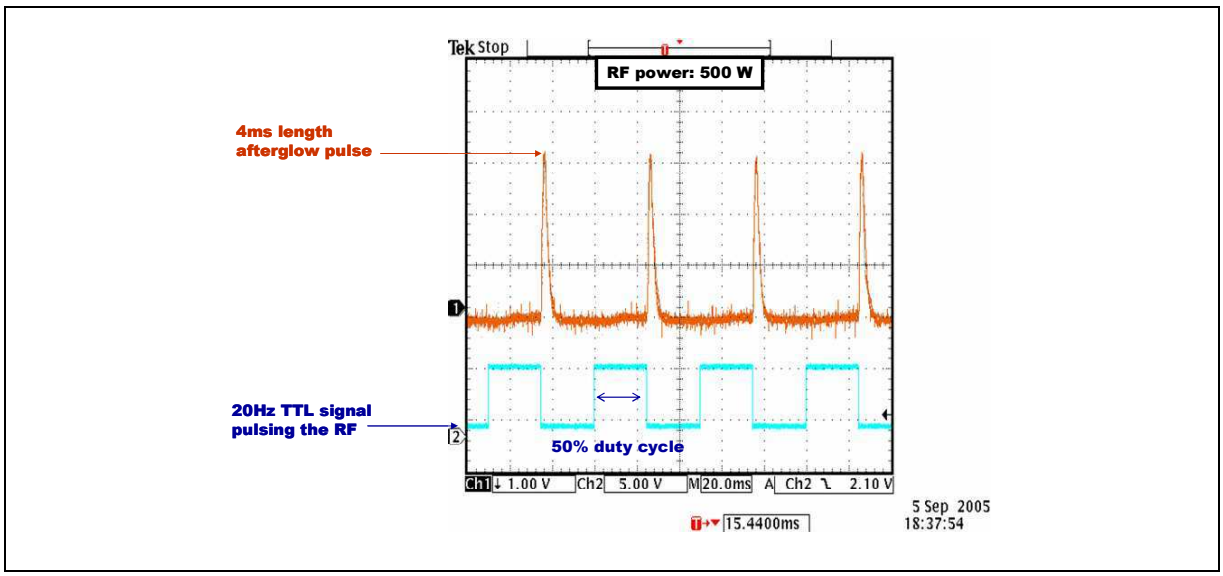

Fig.4 Pulsed mode principle

In pulsed mode, the radiofrequency (RF) is applied for a fixed time. As it is switched off, all ions are extracted as a single pulse or so-called “afterglow” pulse (see Fig.4). Therefore, the confinement time can be reduced to the minimum time necessary for charge breeding, which can go down to 20ms [9] (see Fig.5). The afterglow pulse can last up to 10ms. As a consequence, isotopes living down to 30ms can be studied in the afterglow mode. However, the charge breeding efficiency currently attainable in pulsed mode is half less than the one obtained in continuous mode, i.e. around 2.5% charge breeding efficiency for one charge state. An advantage of this method is the time reference that it gives for the half-life measurement.

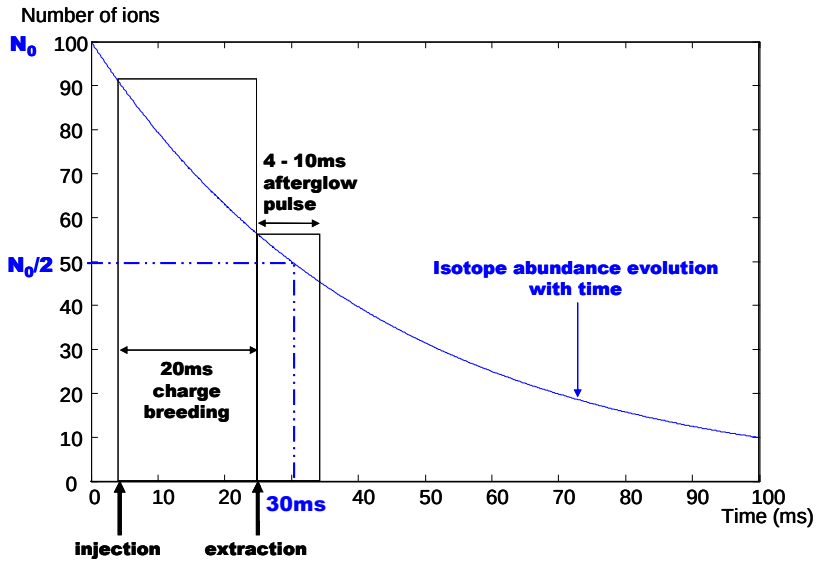

Fig.5 Tuning for short lived isotopes

4. Proposed experiment

4.1. Experimental setup

The experimental setup should consist of a tape station installed after the ECR test bench on GHM, a beta detector (NE102 scintillator cup) in coincidence with at least one HPGe detector for isotopes identification and background estimation, and a neutron counter described in [10].

The detection efficiency can be estimated to 30% for the betas, 19% for the neutrons [10] and about 1% for gamma energies in the range of 500keV-1MeV. The Phoenix Booster can be assumed to have a total injection and charge breeding efficiency of respectively 5% or 2.5% for one charge state in continuous mode or in pulsed mode.

4.2. Cycle description

For each $T_{1/2}$ and P_n measurement, one assumes the following procedure. One starts accumulating ions after the proton pulse, and one stops the accumulation on the tape at the maximum of the release curve with the use of an electrostatic deflector after the analyzing magnet. Doing so, the collection will be optimized according to the release time and expected half life of the isotope in question. The collection will be followed by a counting period $t_{measurement}$ over four half-lives in order to obtain a clean decay curve for both beta and neutrons. The tape is moved afterwards to get rid of the remaining activity. This induces a delay δt_{tape} of approximately 500ms. One has to wait for the next proton pulse coming every $\delta t_{protons}$ of 1.2s to start a new measurement cycle. Therefore, the cycle time t_{cycle} is given in seconds by (eq.1):

(eq.1) $$t_{cycle} = \left(trunc\left(\frac{t_{measurement} + \delta t_{tape}}{\delta t_{protons}}, 0 \right) + 1 \right) \cdot \delta t_{protons}$$

One proton pulse every 2.4s was expected as the highest rate, and each proton pulse has maximum intensity, i.e. 4.8µC.

4.3. Estimated yields and measurement times

We would like to demonstrate first the performances of the cleaning by charge breeding with less exotic GeS^+, $SeCO^+$ and SrF^+ beams, such as $^{82\text{-}83}GeS^+$, $^{86\text{-}89}SeCO^+$ and $^{98\text{-}101}SrF^+$. These have reasonable branching ratios for identification by gamma detection and are therefore easily detectable. The demonstration of feasibility of this method for beam purification, and the yield measurement for such beams would also help other experiments (e.g. REX). For these calibration measurements, we aim for 10% accuracy on the measured $T_{1/2}$ and P_n values.
Then we aim at studying the $^{84\text{-}87}$Ge and $^{90\text{-}92}$Se isotopes by charge breeding them with the continuous mode, and the $^{88\text{-}89}$Ge, ^{93}Se and $^{102\text{-}104}$Sr isotopes with the pulsed mode. For these more exotic isotopes, in order to be competitive with the tabulated values [11] and [12], we aim at measuring $T_{1/2}$ and P_n with uncertainties reduced by a factor 2 compared to former measurements. In case no value was previously reported, we aim at measuring them for the first time with less than 20% uncertainty.

Table 1 gives the current tabulated values of the interesting isotopes for $T_{1/2}$ and P_n with their uncertainties.

The required beams are basically impossible to identify without the proposed additional mass separation after molecular breakup, due to a high background of well-produced isobars such as Ag, Cd, In…etc. The method we propose here was used in a similar way at Oak Ridge National Laboratory (ORNL) [12], where $^{80\text{-}86}GeS^+$ beams were produced with a target-ion source set comparable to what is used at ISOLDE. The molecules were post-accelerated and broken by stripping before undergoing mass and energy separation. Except for the yields measured through this experiment, there are obviously no measured yields available. Therefore we have to estimate probable yields from the measured yields [12], and from available information on less exotic isotopes produced at ISOLDE ([13] and [14]).
Contrary to the experiment at the ORNL, we plan to use a tungsten neutron converter to suppress part of the symmetric fission products. The induced loss factor in the production of the isotope of interest inside a

uranium carbide target was calculated using the fission cross-section that can be found in [15]. Hence, the expected yields for molecular beams of less exotic isotopes such as ^{80}Ge, ^{96}Se and ^{84}Sr could be extrapolated from the available information on measured yields ([13], [14] and [16]). Since the drop of fission yields towards very neutron-rich isotopes of a given element is rather universal, the expected ISOLDE yields could thus be derived (see Table 1).

Then, following the same development as in [17], one calculates the total measurement time. One assumes that a measurement of the total background rate is performed preceding or following the actual $T_{1/2}$ and P_n measurement, with all parameters remaining the same. The internal beta and neutron background is believed to arise mainly from two sources: from the possible neutral radioactive gas diffusing from the GPS target, and from the nearby A/q tails of well-produced isotopes extracted at the same mass as the molecules. For example, the well-produced ^{116}Ag, ^{118}Ag and ^{121}Ag isotopes will be respectively extracted at the same masses as the $^{82}GeS^+$, $^{90}SeCO^+$, $^{102}SrF^+$ molecules, with a yield of about 1E+8ions/s. The factor of suppression for the tails of nearby peaks measured with stable beam in our A/q separation is about 10^{-5}. This leads to a background rate of 20Hz for the betas, and 0.5Hz for the neutrons. For the calculation of the total measurement time, the total background rate was estimated to a maximum of 100Hz for the betas, and 1Hz for the neutrons as in [18].

From the total measurement time and the time for one cycle (eq.1), one can deduce the total time needed for the $T_{1/2}$ measurement and for the P_n measurement, given in Table 1. Detection limits of 1 beta per second and 1 neutron per second were assumed.

Nuclide	$T_{1/2}$	P_n (%)	Estimated yield (ions/μC)	T for $T_{1/2}$	T for P_n
^{82}Ge	4.55s (0.05)	0	2E+03	<1h	-
^{83}Ge	1.85s (0.06)	0	1E+02	< 1 beta/s	-
^{84}Ge	947ms (11)	10.8(6)	6E+00	< 1 beta/s	< 0.1 neutron/s
^{85}Ge	535ms (47)	14 (3)	9E-02	< 1 beta/s	< 0.1 neutron/s
^{86}Ge	200ms (syst)	#?	4E-04	< 1 beta/s	< 0.1 neutron/s
$^{87-89}$Ge	#?	#?	-	-	-
^{86}Se	15.3s (0.9)	0	8E+03	<1h	< 0.1 neutron/s
^{87}Se	5.50s (0.12)	0.20 (4)	5E+03	<1h	3h
^{88}Se	1.53s (0.06)	0.99 (10)	2E+03	<1h	2h
^{89}Se	410ms (40)	7.8 (25)	3E+02	<1h	5h
^{90}Se	#300ms (>300ns)	#?	6E+01	<1h	< 0.1 neutron/s
^{91}Se	270ms (50)	21 (10)	3E+00	< 1 beta/s	< 0.1 neutron/s
$^{92-93}$Se	#?	#?	-	-	-
^{98}Sr	653ms (2)	0.25 (5)	2E+05	<1h	<1h
^{99}Sr	269ms (1)	0.100 (19)	7E+03	<1h	<1h
^{100}Sr	202ms (3)	0.78 (13)	1E+03	<1h	<1h
^{101}Sr	118ms (3)	2.37 (14)	5E+01	2h	< 0.1 neutron/s
^{102}Sr	69ms(6)	5.5 (15)	3E+00	< 1 beta/s	< 0.1 neutron/s
$^{103-104}$Sr	#?	#0 ?	-	-	-

Table 1 Gross properties of the interesting isotopes as in [11] and [12]
The symbol "#" shows estimated values

5. Summary and beam time requirements

Apart from the nuclear astrophysics interest, the measurement of the neutron-rich $^{84+x}$ Ge, $^{90+x}$ Se, $^{102+x}$ Sr isotopes gross properties ($T_{1/2}$, P_n) are so challenging that they were proposed as one of the EURISOL key experiments [19]. The measurement of any of these at an existing facility such as ISOLDE would be a great success. The development of the related techniques of production and separation is in this respect of high

interest. The validation of this purification method is a major step for beam development as it is the only way to determine precisely the yields of the more exotic neutron-rich isotopes.

The study of the gross properties also constitutes a first step towards a deeper investigation of these neutron-rich isotopes through other experiments, for example such as mass measurement or using post-acceleration with REX-ISOLDE.

5.1. Experimental setup

For the experimental setup, a tape station will be needed on the GHM beam line, as well as an HPGe detector, and a neutron counter. The use of the ISOLDE Data Acquisition System is required as well.

5.2. Beam time for on-line measurements

Sufficient beam intensity is expected to perform the $T_{1/2}$ and P_n measurements of the ^{82}Ge, $^{86-90}$Se and $^{98-101}$Sr isotopes with the estimated yields. As the identification and the estimation of the background is a major concern, we require more time than the necessary measurement time reported in the previous section.

For the cases where the count rate was estimated to be below the detection limits, shifts are required for yield investigation, as no precise yield can be measured without the proposed method of additional purification with the ECR.

We require for each element:

- two shifts of preparation, including a yield measurement on less exotic isotopes on the ISOLDE tape station and beam tuning with stable and radioactive ions from GHM to the ECR, four shifts if the conditioning of the afterglow mode is required.
- two shifts of calibration on less exotic but well-known isotopes.
- the shifts necessary for the measurement of the yields for the interesting nuclides, and possibly of $T_{1/2}$ and P_n when the measurement time estimate given in Table 1 is favourable.

Nuclide	Target	Gas leak or mass marker	Ion source	Shifts	Protons averaged current µA	Comments
Ge settings	UC_x ThO_2	^{34}S (mass marker)	MK5	2	-	Beam tuning
^{82}Ge	"	"	"	2	0.3	Calibration
$^{84-87}$Ge	"	"	"	4	1.0-2.0	Investigation of the yields
Se settings	UC_x ThO_2	CO_2	MK5	2	-	Beam tuning
$^{86-89}$Se	"	"	"	2	0.1-2.0	Calibration
^{90}Se	"	"	"	1	2.0	Continuous mode
$^{91-92}$Se	"	"	"	3	2.0	Investigation of the yields
Sr settings	UC_x	CF_4	W surface tungsten	(4)	-	Beam tuning and afterglow conditioning
$^{98-101}$Sr	"	"	"	(2)	1.3-2.0	Calibration
$^{102-104}$Sr	"	"	"	(4)	2.0	Investigation of the yields

Table 2 Beam request for the interesting nuclides
As the Sr case is challenging, no shift is required at this stage

As a summary, a total of 16 shifts is required to perform an extensive study of yields and gross properties for neutron-rich $^{84-87}$Ge and $^{90-92}$Se nuclei on the r-process path for neutron shells between N=50 and N=82.

Preferably, the Ge and Se measurements could be done with the same target, using respectively for the former a mass marker filled with ^{34}S and for the latter a gas leak of CO_2. Moreover, we know from the release of neutron deficient Se [20] that oxide targets would allow a more rapid release and, hence, higher beam intensities. This conclusion might also be applied to the production of Ge, i.e. a test of a ThO_2 target could give significantly higher yields compared to the conservative estimates used in the previous section for UC_x target. This study would strongly benefit from such a target, as the number of accessible isotopes could be significantly increased.

At this stage, no shift is required for the study of $^{102-104}$Sr because these measurements appear quite challenging considering the technique to be used, i.e. the pulsed mode, or so-called afterglow, which still needs to be studied. For the same reason, no shift is required for $^{88-89}$Ge and ^{93}Se either. However, as Sr beam developments are planned for the beginning of the year, one might consider the possibility of performing some test measurements with the ECR in parasitic mode. A beam request might be formulated later.

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